

TRANSFORMING RICE HUSK INTO A HIGH-ADDED VALUE PRODUCT: POTENTIAL FOR PARTICLEBOARD PRODUCTION

TRANSFORMAÇÃO DA CASCA DE ARROZ EM UM PRODUTO DE MAIOR VALOR AGREGADO: POTENCIAL PARA A PRODUÇÃO DE PAINÉIS PARTICULADOS

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ABSTRACT

This work aimed to evaluate the quality of particleboards made with different formulations, varying adhesive content and rice husk (*Oryza sativa L.*) proportions added to *Cunninghamia lanceolata* wood. Three adhesive contents (6, 9 and 12%) and three proportions of rice husk added to wood (25, 50 and 75%) were combined resulting in 9 formulations of particleboards. In order to analyze the influence of each raw material in the final quality of the boards, their chemical composition was determined, including extractive, ash, lignin and holocellulose contents. Regarding the particleboards, the following physical and mechanical properties were evaluated: water absorption (WA), thickness swelling (TS), internal bonding (IB), parallel compression (PC), modulus of elasticity (MOE) and modulus of rupture (MOR) in static bending. For both mechanical and physical properties, average values showed that lower rice husk proportions and higher adhesive contents induced to better performance. Low lignin and high ash contents found in rice husks negatively influenced particleboard quality. Among the formulations tested, the inclusion of 25% of rice husk and 9% urea-formaldehyde presented the best results. Particleboards produced in this work are potential alternatives for general applications in dry conditions that do not require high mechanical resistance. Nevertheless, the process and formulations need to be adjusted in order to reach the requirements for more demanding structural applications.

Keywords: *Cunninghamia lanceolata*; lignocellulosic materials; internal bonding; adhesive content.

RESUMO

O objetivo deste trabalho foi avaliar a qualidade de painéis particulados produzidos com diferentes formulações, variando percentagens de adesivo e casca de arroz (*Oryza sativa L.*) adicionadas à madeira

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de *Cunninghamia lanceolata*. Três teores de adesivo (6, 9 e 12%) e três proporções de casca de arroz (25, 50 e 75%) foram combinados resultando em 9 formulações. Para analisar a influência de cada matéria-prima na qualidade final dos painéis, sua composição química foi determinada de acordo com os teores de extrativos, componentes minerais, lignina e holocelulose. Os painéis aglomerados foram avaliados pelas seguintes propriedades físicas e mecânicas: absorção de água (AA), inchamento em espessura (IE), ligação interna (LI), compressão paralela (CP), módulo de elasticidade (MOE) e módulo de ruptura (MOR) na flexão estática. Os valores médios das propriedades físicas e mecânicas demonstraram que menores teores de casca de arroz e maiores teores de adesivo ocasionaram melhor desempenho dos painéis. O baixo teor de lignina e alto teor de cinzas da casca de arroz influenciaram negativamente a qualidade dos painéis. Dentre as formulações avaliadas, painéis com inclusão de 25% de casca de arroz e 9% de ureia-formaldeído apresentaram os melhores resultados. Os painéis produzidos neste trabalho são alternativas potenciais para aplicações gerais em condições secas que não demandem alta resistência mecânica. No entanto, o processo e as formulações necessitam ser ajustados para aplicações estruturais mais exigentes.

Palavras-chave: *Cunninghamia lanceolata*; materiais lignocelulósicos; ligação interna; teor de adesivo.

INTRODUCTION

The modern society, with its high population density, sophisticated industries and intensive methods of agriculture, produces wastes in an increasing scale (ANTONIOLLI et al., 2009). Since the basic principle of particleboard production is that any lignocellulosic material may be used as raw material, the utilization of wastes in those industries may be an interesting alternative from both economic and environmental points of view. The demand for alternative lignocellulosic resources to substitute wood in particleboard production has increased in the last years and led to many researches on the field (CIANNAMEA et al., 2010; FREIRE et al., 2015; MELO et al., 2009). Materials such as wood harvesting wastes, husks, annual plants, plant wastes, wastes from cellulose industry, recycled paper and other may supply this demand (AKGUL; ÇAMLIBEL, 2008). This practice ensures value adding to those materials and prevents their improper polluting disposal.

Among possible alternative raw materials for particleboard production, rice husk should be taken into account. Brazil produced 12.16 million tons of rice in 2014, which placed the country as the ninth largest producer (FAO, 2014). From this production, 20% corresponds to residual husk (FOLETTTO et al., 2005). Rice husk is a cellulose based fiber that may be used in the production of panels and composites (NDAZI et al., 2007). Several papers verified the feasibility of using rice husk in particleboard production (AJIWE et al., 1998; CINNAMEA et al., 2010; GERARDI et al., 1998; JONHSON; YUNUS, 2009; LEE et al., 2003; LEIVA et al., 2007; NDAZI et al., 2007; NWACHUKWU, 2007; OSARENMWINDA; SAMPATHRAJAN et al., 1992).

Although agricultural panels are environmentally friendly, in some cases the panels present poor mechanical and water resistance, which limits their commercialization. For this reason, the mixture of agricultural wastes and wood may be an alternative solution.

Cunninghamia lanceolata (Lamb.) Hooker f. (*Cunninghamia*, Taxodiaceae) is one of the most important commercial softwood plantation species in southern China. The most recent national forestry survey indicates that the current distribution area of *Cunninghamia lanceolata*, commonly named Chinese fir, plantations is about 9.22 million ha (YIN et al., 2010). The wood obtained from this species presents high ratio of heartwood and very good durability due its extractives (SONG et al., 2011; WANG et al., 2011). No papers concerning the use of *Cunninghamia lanceolata* wood in particleboard production can be found in the literature until the moment.

Therefore, this work aimed to evaluate the physical and mechanical resistance of particleboards made with different formulations, varying adhesive content and rice husk proportion added to *Cunninghamia lanceolata* wood.

MATERIAL AND METHODS

Materials

The 18-year-old *Cunninghamia lanceolata* wood was supplied by Melhoramentos SA industry of Monte Verde, southern Minas Gerais state, Brazil. For particleboard production, veneering wastes were milled to produce sliver particles using a 6.14 mm sieve fastened to a hammer mill. The particles were screened through a 0.6 mm sieve to exclude the fine material.

Rice husks were provided by the rice processing industry 'Pereira e Teodoro Ltda', located in Lavras, southern Minas Gerais state, Brazil.

The basic density of the wood was determined according to NBR 11941 (ABNT, 2003) standard, while rice husk density was determined by water displacement method (AZZINI et al., 1981). Total extractives and lignin contents were determined in accordance with M3/69 (ABTCP, 1974a) and M70/71 (ABTCP, 1974b) standards, respectively. The ash contents of the wastes were determined according to standard M11/77 (ABTCP, 1974c) using a muffle furnace.

Urea-formaldehyde adhesive was purchased from Derquin industry, Curitiba, Paraná, Brazil.

Particleboard formulation and processing

Wood particles and rice husks with 3% initial moisture content were blended with urea-formaldehyde (UF), with 62.37 wt% solids, and 1wt% wax emulsion using a rotatory drum blender for 10 min at room temperature. The mixtures were subsequently molded into particleboards in a 48.5 X 48.5 wood mold. Nine formulations were tested correspondent to variations in adhesive content and proportions of lignocellulosic materials (Table 1).

Press time, temperature and pressure were 8 min, 433 K, and 3.92 MPa. Target bulk density and thickness for all boards were 0.700 g cm⁻³ and 1.5 cm, respectively. The obtained boards were trimmed to avoid edge effects. Three replicates for each formulation of particleboards presented were produced.

Particleboard evaluations

The samples were stabilized in an acclimatized room (298 ± 1 K and 65 ± 3%). The following physical and mechanical properties were evaluated: water absorption after immersion for 02h (WA_2h) and 24h (WA_24h) (two 150 x 150 mm specimens per particleboard); thickness swelling after immersion for 02h (TS_2h) and 24h (TS_24h) (two 150 x 150 mm specimens per particleboard); measured density

TABLE 1: Descriptors of the experimental particleboards.

TABELA 1: Delineamento experimental dos painéis particulados.

Formulations	Proportions of lignocellulosic materials (wt %)		UF content (wt%)
	Rice husk	<i>Cunninghamia lanceolata</i>	
RC75/6	75	25	
RC50/6	50	50	6
RC25/6	25	75	
RC75/9	75	25	
RC50/9	50	50	9
RC25/9	25	75	
RC75/12	75	25	
RC50/12	50	50	12
RC25/12	25	75	

(two 150 x 150 mm specimens per particleboard); modulus of elasticity (MOE) and modulus of rupture (MOR) in three point static bending (four 250 x 50 mm specimens per particleboard); parallel compression (PC) (four 25 x 100 mm specimens per particleboard); and internal bond (IB) (six 50 x 50 mm specimens per particleboard). Physical properties, IB and PC were determined in accordance with ASTM D 1037-100 (ASTM, 2006) standard procedures, while static bending test was performed according to DIN 52362 (DIN, 1982) procedure. For mechanical tests, a universal testing machine (EMIC DL-30000) was used.

Data analysis

The experiment was arranged in a completely randomized design in a 3 X 3 (3 rice husk proportions and 3 adhesive contents) factorial scheme. The data was submitted to variance analysis to verify possible interactions between factors. Average values were compared by Tukey test at 5%.

RESULTS AND DISCUSSION

Raw materials characterization

The average basic density found for *Cunninghamia lanceolata* wood was 0.426 g cm^{-3} , while for rice husk the value found by water displacement method was 0.267 g cm^{-3} . Figure 1 depicts the chemical composition of the raw materials studied.

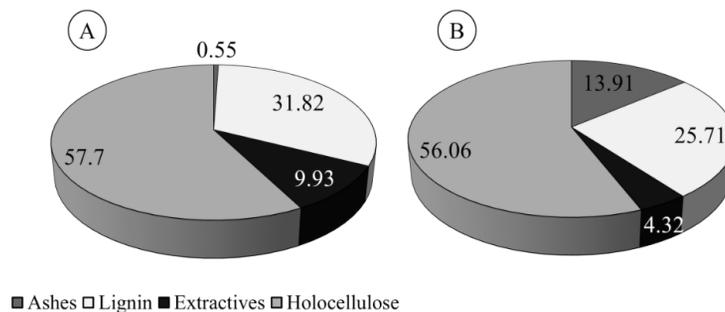


FIGURE 1: Chemical composition of lignocellulosic raw materials: A) *Cunninghamia lanceolata* wood; B) Rice husk.

FIGURA 1: Composição química dos materiais lignocelulósicos: A) Madeira de *Cunninghamia lanceolata*; B) Casca de arroz.

Lignin content was far higher in *Cunninghamia lanceolata* wood than in rice husk which is advantageous for particleboard production. Since lignin is a natural wood binder, TS and WA values of particleboards made from high lignin content materials are lower because of the improved bond formation between particles during mat forming process (KHEDARI et al., 2004; SELLERS et al., 1988). In addition, lignin acts as a stiffener of cellulose microfibrils and appears to prevent or limit perpendicular movement to the grain resulting in higher mechanical strength (SWEET; WINANDY, 1999).

Lower extractive content was detected for rice husks in comparison to the studied wood. Extractives are relevant for wood-adhesive interaction from both chemical and physical points of view. Their high content may decrease surface reaction of the lignocellulosic materials, besides blocking adhesive entering by superficial concentration in high temperatures used during pressing (FRIHART; HUNT, 2010). However, it should be mentioned that besides quantitative variations, extractives also vary in chemical nature and depend on species and environmental conditions (BALLESTEROS et al., 2011; BRÉMAUD et al., 2011; KASSENEY et al., 2011). Therefore, their effect on particleboard production may also vary. For instance, although they exhibited higher extractive contents in this work, rice husks are an important source for wax (SAMUDITHA et al., 2009). Those components possibly hinder adhesive entrance in the particle during pressing.

Ash content was much higher in the rice husks. Those wastes are known for their high silica content which certainly composed the major proportion of the ash determined. Silica occurs in the cell walls in virtually all aerial parts of rice plant, and is most abundant in the husk (PARK et al., 2003). Ashes from calcination of rice husk deep bedding contain about 42% of silica oxide (DI CAMPOS et al., 2008). The presence of higher concentrations of this component and waxes in rice husk than those of wood may hide reactive sites for adhesion with polar adhesives, affecting panel bond resistance (NDAZI et al., 2007; PARK et al., 2003). It was reported that panels that had silica removed from their cell walls showed better mechanical and physical performance (CINNAMEA et al., 2010).

Mechanical properties

The results of the variance analyses for all mechanical properties are depicted in Table 2. As no significant interaction was verified between rice husk proportion and adhesive content for any of the mechanical properties, the factors were evaluated separately (Figures 2, 3, 4 and 5). Increases in adhesive content resulted in better mechanical resistance. This tendency may be attributed to the higher adhesive amount available per particle which leads to strong contact between them. Moreover, average values showed that lower rice husk proportions induced better performance to the obtained particleboards, while adhesive content affected MOE, MOR and IB, but not PC.

The specifications stipulated by EN 312-3 (EN, 2003) for boards Type P5, P4, P3 and P2 do not allow the commercialization of any of the formulations of the proposed particleboards. However, the commercialization of general purpose boards for use in dry conditions (P1) only require minimum values of

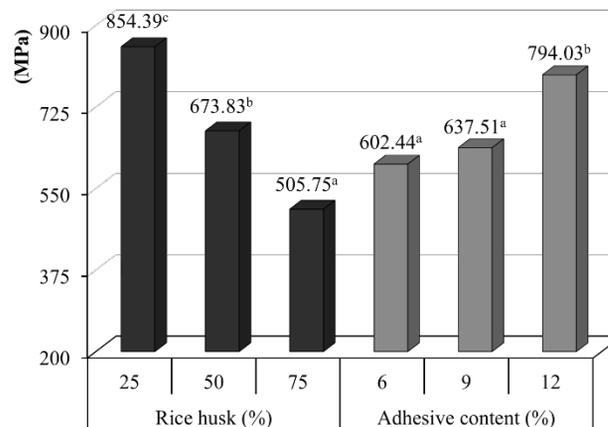


FIGURE 2: Average values of MOE. Averages followed by the same letter do not differ by Tukey test at 5%.
 FIGURA 2: Valores médios de módulo de elasticidade. Valores seguidos de mesma letra não diferem entre si pelo teste Tukey a 5% de significância.

TABLE 2: Results of the variance analyses for mechanical properties.

TABELA 2: Resultados da análise de variância para as propriedades mecânicas.

Variation source	Calculated F			
	MOE	MOR	PC	IB
Rice husk	38.984*	26.619*	55.234*	137.149*
Adhesive	13.345*	6.888*	2.554 ^{ns}	6.040*
Rice husk*Adhesive	1.544 ^{ns}	1.305 ^{ns}	0.921 ^{ns}	0.280 ^{ns}
CVe (%)	12.36	18.50	15.68	18.43

where in: CVe: experimental coefficient of variation; e*: significant at 5% level; e^{ns}: non-significant at 5% level.

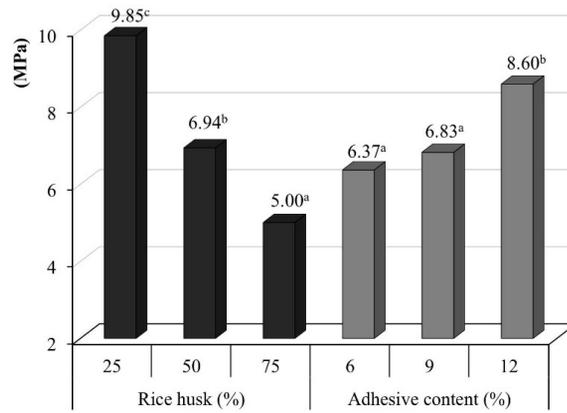


FIGURE 3: Average values of MOR. Averages followed by the same letter do not differ by Tukey test at 5%.

FIGURA 3: Valores médios de módulo de ruptura. Valores seguidos de mesma letra não diferem entre si pelo teste Tukey a 5% de significância.

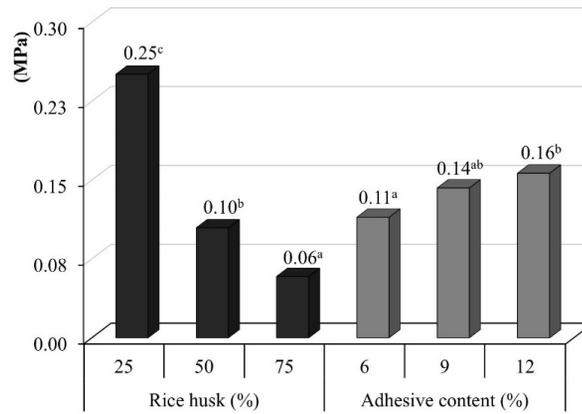


FIGURE 4: Average values of IB. Averages followed by the same letter do not differ by Tukey test at 5%.

FIGURA 4: Valores médios de ligação interna. Valores seguidos de mesma letra não diferem entre si pelo teste Tukey a 5% de significância.

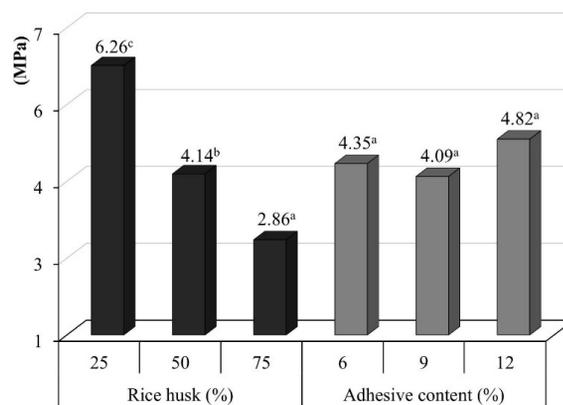


FIGURE 5: Average values of PC. Averages followed by the same letter do not differ by Tukey test at 5%.

FIGURA 5: Valores médios de compressão paralela. Valores seguidos de mesma letra não diferem entre si pelo teste Tukey a 5% de significância.

MOR (11.5 MPa) and IB (0.24 MPa). Therefore, 25% rice husk particleboards presented adequate average value of IB, independently of the adhesive content. The same was not observed for MOR. Decreases in static bending properties due to increases in rice husk levels in particleboard composition has already been observed in the literature (MELO et al., 2009).

ANSI A208.1 (ANSI, 1999) standard demands IB, MOE and MOR values of 0.10, 550 and 3 MPa, respectively for door core application (LD-1). Panels produced with 25 and 50% could be commercialized for this application, independently of the adhesive content. According to mechanical test results, all formulations proposed for particleboard production failed to reach minimum requirements of MOE (17.16 MPa), MOR (3089.10 MPa) and IB (0.41) stipulated by CS 236-66 (CS, 1968) standard.

In general, the poor IB may be considered the main reason for lowering all the other mechanical properties, which are strongly related to high contact between particles. Azizi et al. (2011) attributed low MOE, MOR and IB values to weak interfacial interaction between wheat straw and urea-formaldehyde adhesive.

For many practical reasons, MOE, MOR and IB are among the technological properties of engineering wood products which may ensure their suitability as structural elements (HEIN et al., 2011). The results presented in this work suggest that the inclusion of rice husk in particleboards is unlikely to result in high-strength materials, hence the goal should be to develop boards to be commercialized for internal utilizations.

Physical properties

There was no significant difference in target density between treatments and the average values ranged from 0.640 to 0.700 gcm⁻³ (calculated $F = 2.486$). The results of the variance analyses for the other physical properties are depicted in Table 3. As no significant interaction was verified between rice husk proportion and adhesive content for any of the physical properties, the factors were evaluated independently.

The effects of rice husk proportions and adhesive contents on physical properties are shown in Figures 6 and 7, respectively. Particleboards with incorporation of 75% rice husk exhibited the highest WA and TS values at both 02 and 24h water immersion, followed by particleboards with 50 and 25% rice husk incorporation. Once water penetrates, it may diffuse into the raw materials through amorphous regions of cellulose, which may be considered the main reason for WA (CIANNAMEA et al., 2010). This result is in accordance with the chemical composition of rice husk that may interfere in bond quality and physical properties. Regarding TS, different lignocellulosic materials present different chemical and anatomic properties which will affect their water uptake and swelling. The amount of the stress and their possible effects on TS depend on ability of the adhesive to resist alternating swellings between adjacent particles (MALANIT et al., 2011).

Particleboards made with 12% urea-formaldehyde presented an improvement in WA and TS after 02h in comparison with those made with 6% urea-formaldehyde. 9% urea-formaldehyde particleboards presented values of water resistance after 24h significantly equal to those found for other contents. On the other hand, 12% urea-formaldehyde particleboards exhibited the lowest TS after 24h.

The increase of water resistance with increases in adhesive content may be mainly attributed to

TABLE 3: Results of the variance analyses for physical properties.

TABELA 3: Resultados da análise de variância para as propriedades físicas.

Variation sources	Calculated F			
	WA_2h	WA_24h	TS_2h	TS_24h
Rice husk	110.190*	115.630*	166.318*	225.485*
Adhesive	5.038*	4.167*	6.388*	13.184*
Rice husk*Adhesive	2.189 ^{ns}	0.426 ^{ns}	2.024 ^{ns}	2.915 ^{ns}
CVe (%)	13.63	9.17	13.90	10.27

where in: experimental coefficient of variation; e*: significant at 5% level; e^{ns}: non-significant at 5% level.

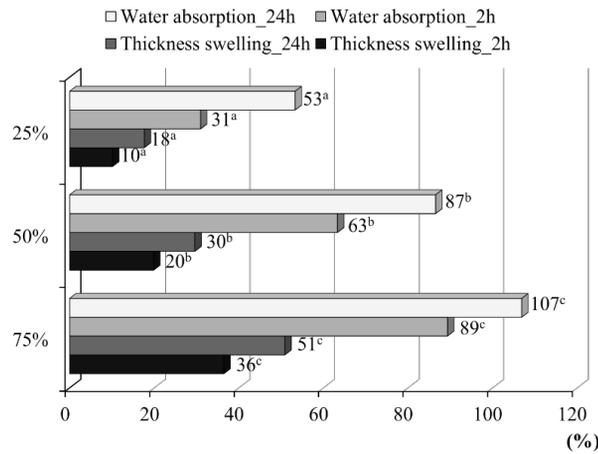


FIGURE 6: Average values of physical properties for rice husk amount factor. Averages followed by the same letter do not differ by Tukey test at 5%.

FIGURA 6: Valores médios das propriedades físicas para o fator casca de arroz. Valores seguidos de mesma letra não diferem entre si pelo teste Tukey a 5% de significância.

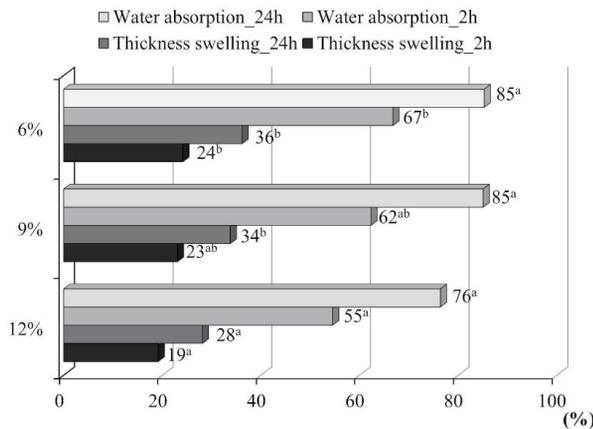


FIGURE 7: Average values of physical properties for adhesive content factor. Averages followed by the same letter do not differ by Tukey test at 5%.

FIGURA 7: Valores médios das propriedades físicas para o fator teor de adesivo. Valores seguidos de mesma letra não diferem entre si pelo teste Tukey a 5% de significância.

the higher adhesive amount per particle, which may improve the glue bonding and promote higher contact between particles. Similar trends of dimensional instability in relation to adhesive content for agriculture particleboards have already been reported in the literature (GRIGORIOU et al., 2000; MENDES et al., 2010).

The EN 312-3 (EN, 2003) standard for TS after 24h requires values below 13%. Thus, none of the particleboard formulations resulted in TS below the maximum stipulated by this standard. ANSI A208.1 (ANSI, 1999) only specifies TS maximum value for boards applied in manufactured home decking, which should be below 8%; hence the formulations proposed in this work do not meet the requirements for this application, but they have no restrictions for other applications described in the standard. Based on CS 236-66 (CS, 1968) standard, particleboards should have a maximum TS value after 24h of 25%. The average TS values of particleboards made with 9 and 12% adhesive contents and 25 and 50% coffee husk proportion met the requirement mentioned above.

TS could be affected by several factors such as insufficient resin content and distribution, poor

compatibility between substrate and adhesive, and adhesive properties (WANG; SUN 2002). When the bonding strength resulting from UF adhesive is not strong, a larger amount of water is able to penetrate the weakly bonded particleboard structure (CIANNAMEA et al., 2010). Therefore, high TS values were certainly influenced by low IB values showed in the discussion above.

CONCLUSIONS

The chemical composition of rice husk strongly affected the quality of the particleboards produced in this work, since higher proportions of this waste in the formulation decreased physical and mechanical strength. Poor bonding provided by rice husk in particleboards was attributed to high extractive and ash contents and low lignin content.

Overall analysis showed that the best formulation found for production of rice husk/*Cunninghamia lanceolata* particleboards, was the inclusion of 25% of the agricultural waste and 9% urea-formaldehyde.

Particleboards produced in this work are potential alternatives for general applications in dry conditions that do not require high mechanical resistance. Nevertheless, the process and formulations need to be adjusted in order to reach the requirements for more demanding structural applications.

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